Spin excitations in a single La_2CuO_4 layer

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The dynamics of $S = \frac{1}{2}$ quantum spins on a 2D square lattice lie at the heart of the mystery of the cuprates [1–7]. In bulk cuprates such as La_2CuO_4, the presence of a weak interlayer coupling stabilizes 3D Néel order up to high temperatures. In a truly 2D system however, thermal spin fluctuations melt long range order at any finite temperature [8]. Further, quantum spin fluctuations transfer magnetic spectral weight out of a well-defined magnon excitation into a magnetic continuum, the nature of which remains controversial [6, 9–11]. Here, we measure the spin response of *isolated one-unit-cell thick layers* of La_2CuO_4. We show that coherent magnons persist even in a single layer of La_2CuO_4, with no evidence for resonating valence bond (RVB)-like spin correlations [11–13]. Thus these excitations are well described by linear spin wave theory (LSWT). We also observe a high-energy magnetic continuum in the isotropic magnetic response. This high-energy continuum is not well described by 2 magnon LSWT, or indeed any existing theories.

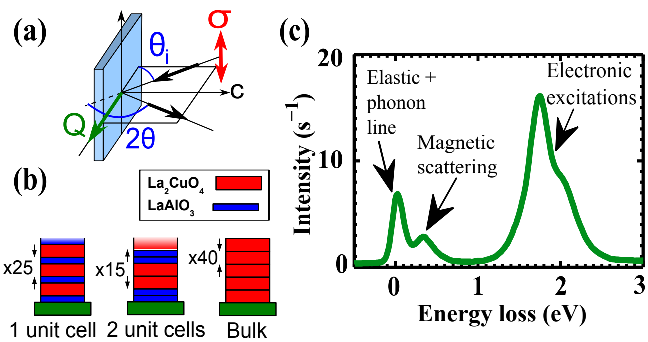

FIG. 1. (a) The experimental scattering geometry. The 931 eV σ polarized x-rays are incident at an angle θ_i and are scattered through a fixed angle $2\theta = 130°$. Large $\boldsymbol{Q}$ corresponds to near-grazing incidence ($\theta_i \to 0$). (b) The multilayer films studied, composed of 13.2 Å La_2CuO_4 layers (red blocks) and 3.8 Å $LaAlO_3$ (blue blocks). We label the films, based on the thickness of La_2CuO_4, as 1 unit cell, 2 unit cell and and bulk. The arrows denote the repeat unit of the films (×25, ×15, ×40). (c) A representative RIXS spectrum of the 1 unit cell La_2CuO_4 film ($25\times[LaAlO_3 + La_2CuO_4]$) at $\boldsymbol{Q}$= (0.77π,0) identifying the main spectral features: the elastic and phonon scattering around zero energy transfer, the magnetic scattering around 300 meV and the electronic (dd) excitations from 1 to 3 eV.

The simplest model for describing the magnetic excitations of undoped cuprates is LSWT [14]. Coherent transverse magnetic excitations correspond to spin waves – magnons – with a well-defined energy; whereas longitudinal magnetic excitations result in a high-energy continuum of multi-magnons. While measurements of the long wavelength magnetic excitations of La_2CuO_4 [15] can be understood in terms of a renormalzied classical model [16], the short range correlations remain controversial [6, 9–12, 17] as quantum fluctuations can transfer spectral weight out of the magnon peak into a high-energy continuum. Furthermore, the magnetic excitation spectrum of a 2D one-unit-cell-thick La_2CuO_4 layer, where spin fluctuations are expected to be enhanced, has not been measured. This is because most of what we know about the spin excitation spectrum of the cuprates has come from inelastic neutron scattering. Unfortunately, such experiments require large samples and are often challenging at high energy transfers. In recent years, however, resonant inelastic x-ray scattering (RIXS) has achieved sufficient resolution to access magnetic excitations [7, 18–20] and RIXS is well suited to measuring high-energy magnetic excitations in the range 100-1000 meV. Furthermore, the high sensitivity of the technique allows us to look at nanostructured samples and this in turn opens up the exciting possibility of measuring the spin response of a single La_2CuO_4 layer for the first time.

We performed RIXS measurements on bulk and single layer La_2CuO_4 films at 15 K using the scattering geometry shown in Fig. 1(a). The sample was rotated about the vertical axis to vary $\boldsymbol{Q}$, the projection of the total scattering vector in the ab-plane. To provide sufficient scattering volume of isolated La_2CuO_4 layers, we prepared heterostructures based on 1 unit cell (uc), i.e. 2 CuO_2 planes, thick layers of La_2CuO_4 and $LaAlO_3$. The samples are depicted in Fig. 1(b) as 1uc =[1uc $LaAlO_3$ + 1uc La_2CuO_4]×25, 2uc =[2uc $LaAlO_3$ + 2uc La_2CuO_4]×15, and bulk =[1uc La_2CuO_4]×40. Muon spin rotation studies imply that the La_2CuO_4 layers are well isolated from one another (see supplementary materials) and the RIXS results (discussed later) show that the correlated patches of spins are randomly orientated as expected for an isolated La_2CuO_4 layer.

The RIXS spectra of the three samples were measured from (0.14π,0) to (0.8π,0) and (0.1π,0.1π) to (0.6π,0.6π). Figure 1(c) plots a representative spectrum collected at $\boldsymbol{Q}$= (0.77π,0). We observe a peak corresponding to elastic scattering around zero energy transfer, which also contains a shoulder at low energy transfers (up to ∼90 meV) due to phonon scattering. From 200-800 meV we observe magnetic scattering and in the 1-3 eV window we identify electronic *dd*-excitations.

The elastic and phonon peak in the spectra were fit using Gaussian functions and subtracted from the data, in order to isolate the magnetic scattering. In the bulk film the response is dominated by a dispersing magnon peak (see Fig. 2(a)), along with additional scattering extending out to higher energies. As $\boldsymbol{Q} \rightarrow 0$ the specular reflection from the sample surface overwhelms any magnetic signal. For the case of the 1uc and 2uc films, shown in Fig. 2(b) and (c), the peak intensity is suppressed and the peak width is broadened with additional weight at high energies.

Figure 2(d) plots the dispersion of the peak in the magnetic response of the three films. We see that the 1uc and 2uc films still display a coherent magnon peak, with the same dispersion as in the bulk. For comparison we also plot neutron scattering results from bulk La_2CuO_4 [6]. These are in excellent agreement along (0,0) to $(\frac{\pi}{2},\frac{\pi}{2})$. Along (0,0) to ($\pi$,0) our results appear slightly higher in energy than Ref. [6]. We attribute this to the fact that in the present case we are recording the median energy of the asymmetric peak, rather than the peak energy position, as is the case in Ref. [6]. This difference has a bigger effect along the (π,0) direction because of the increased high-energy tail in this direction, a fact we will return to later.

Figure 2(d) shows the principal result of this paper: Even in La_2CuO_4 layers only a single unit cell thick, a coherent, bulk-like magnon is a reasonable description of the magnetic excitations. Significantly, the dispersion is very similar to that of bulk La_2CuO_4. Thus, even though LSWT is based on an ordered Néel state, it continues to provide a reasonable description of the spin response of a one-unit-cell-thick La_2CuO_4 layer. This is despite the fact that the Néel ordered state is predicted to be suppressed in the limit of a single CuO_2 plane [16].

This is an important result. Recent muon spin rotation data seem to indicate that thin La_2CuO_4 layers are dominated by quantum fluctuations [17]. Indeed, recent calculations suggest that quantum fluctuations may be enhanced in La_2CuO_4 because of frustrated higher order hopping [21]. Therefore, a key unanswered question is whether a single unit cell layer of La_2CuO_4 hosts a more RVB-like state, or whether it obeys the expectations of the Heisenberg model with renormalized classical correlations? RVB-like models, shown as the dotten line in Fig. 2(d), predict a much lower energy at (π,0) than at $(\frac{\pi}{2},\frac{\pi}{2})$ [13]. In contrast, Fig. 2(d) shows no such downturn and further our data imply the presence of similar magnetic correlations in single layer and in bulk La_2CuO_4. Thus, our results strongly support the second scenario.

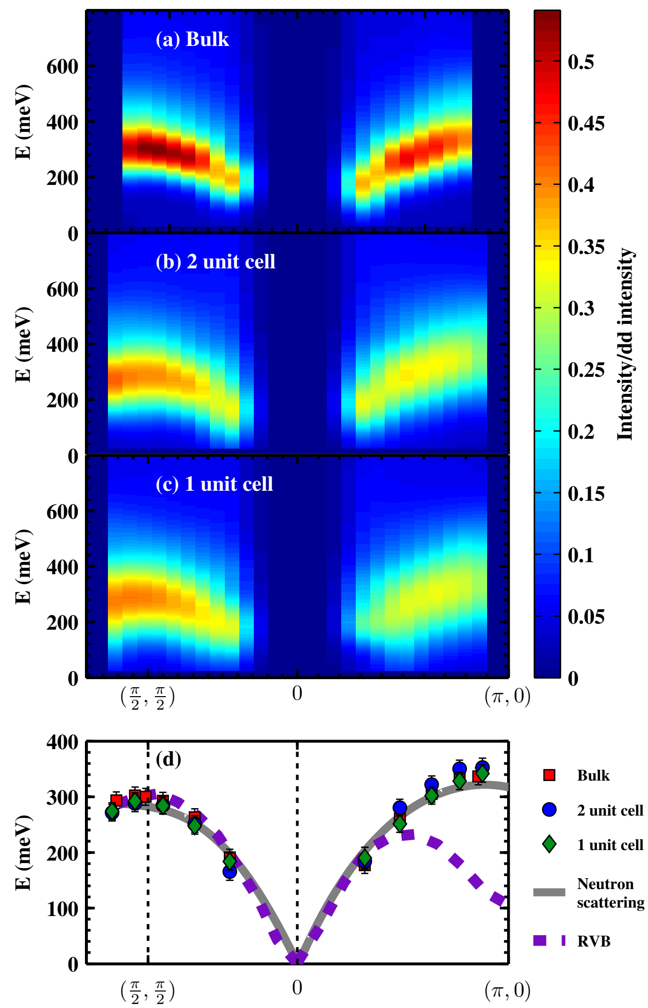

FIG. 2. (a)-(c) The magnetic RIXS scattering intensity along the high symmetry lines in the BZ for: (a) Bulk film, (b) 2 unit cell (15×[2 $LaAlO_3$ + 2 La_2CuO_4]) and (c) 1 unit cell (25×[$LaAlO_3$ + La_2CuO_4]) samples. (d) The peak energy dispersion in bulk (red □), 2 unit cell (blue ∘) and 1 unit cell La_2CuO_4 (green ⋄). The solid gray line is the result from neutron scattering measurements of bulk La_2CuO_4 [6]; the dotted purple line are calculations for an RVB model [13].

More specifically, our single layer data are consistent with the renormalized classical expectation for the Heisenberg model: Even though thermal fluctuations

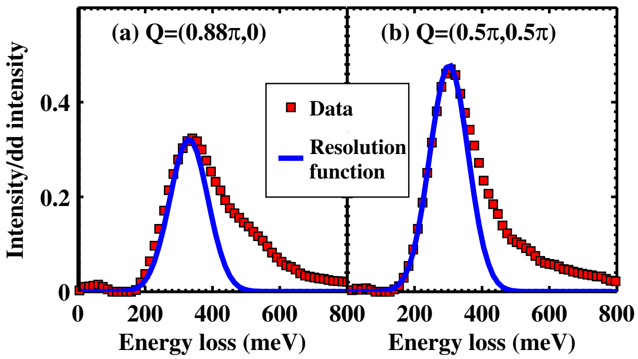

FIG. 3. The measured magnetic spectral weight (red □) in bulk La_2CuO_4 at (a) $\boldsymbol{Q}$=(0.88π,0) and (b) $\boldsymbol{Q}$=(0.5π,0.5π). The blue line is a resolution-limited Gaussian at the single magnon energy. The statistical error bars are smaller than the point size.

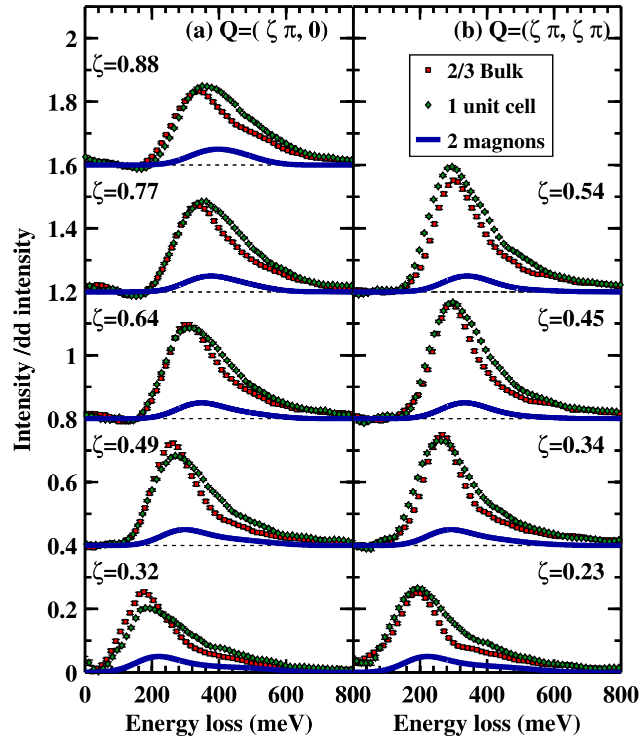

FIG. 4. A comparison between $\frac{2}{3}$ of the bulk La_2CuO_4 response (red □) and the 1 unit cell ($25\times[LaAlO_3 + La_2CuO_4]$) response (green ◇) along (a) ($\zeta\pi$, 0) and (b) ($\zeta\pi$, $\zeta\pi$). The difference between these spectra corresponds to the longitudinal magnetic response, to be compared to the two magnon $S^{zz}(\boldsymbol{Q},\omega)$ calculation (blue line) [21]. As it is not currently possible to measure absolute magnetic scattering intensities with RIXS, the calculation is shown with an arbitrary intensity.

suppress long range order at any finite temperature in a true 2D system, as the temperature is lowered, the correlation length increases exponentially in J/k_BT [16] the spin wave lifetime likewise increases as correlation length divided by spin wave velocity [22], and the system mimics a Néel state in its response. Neutron scattering studies of the nearest-neighbour material $Cu(DCOO)_2{\cdot}4D_2O$ have shown a small quantum correction to the magnon energy at (π,0) [11, 23]. In contrast our data show a 40 meV (13%) higher energy at (π,0) than at ($\frac{\pi}{2}$,$\frac{\pi}{2}$), thereby confirming the neutron measurements on bulk La_2CuO_4 [5, 6]. We conclude that even in the single La_2CuO_4 layer, the quantum corrections to the dispersion are small and hidden by a larger zone boundary dispersion due to the longer ranged interactions resulting from higher order hopping terms [5, 6, 19, 21].

We are now in a position to study the 2D $S = \frac{1}{2}$ response in detail. To do so, we first consider the bulk response, Fig. 3. A continuum of magnetic excitations is observed above the single magnon energy extending out to 800 meV, beyond which the tail of the *dd* excitations makes it hard to determine the origin of the scattering. Similar high-energy continua are seen at all momenta, as is evident from Fig. 2. We note that neutron scattering also sees high-energy magnetic scattering up to the maximum measured energy transfer of 450 meV [6]. In Fig. 3, we see that this scattering is anisotropic; this high-energy weight is stronger near (π,0) than at ($\frac{\pi}{2}$,$\frac{\pi}{2}$) relative to the magnon peak.

In first-order LSWT, the magnons are fluctuations transverse (T) to the spin quantization axis, and result in a resolution-limited magnon peak, with no weight at higher energies. Quantum fluctuations shift weight to a higher-energy continuum, which within SWT is longitudinal (L) two-magnon scattering. In addition, Quantum Monte Carlo calculations [9] and neutron scattering [22] for a nearest-neighbour Heisenberg antiferromagnet also indicated a transverse continuum especially around (π,0). It has been speculated that this transverse continuum can in part be described as fractional spinon-like excitations [10, 12]. There is also the possibility that part of this transverse continuum is three magnon processes, which RIXS may be sensitive to [24, 25].

With this description of the bulk scattering in hand, we now turn to the 1uc film for which we expect there to be no broken spin rotational symmetry. Comparing Fig. 2(a) and (c) we find that the 1uc film shows a smaller peak intensity than the bulk and more spectral weight at high energies. As shown in Refs. [25–27] Cu L-edge RIXS, with σ polarized incident light, is dominated by the out-of-plane c-axis magnetic response. In the bulk sample, magnetic moments order in the plane along the (110) axis, defining the spin quantization axis. Thus, here RIXS measures the transverse response T. If the isolated La_2CuO_4 layers in a 1uc film do not order, one would expect the domains of locally correlated spins to be randomly orientated either in our out of the CuO_2 planes. In this case, one then measures the isotropic response $\frac{2}{3}T + \frac{1}{3}L$. Figure 4 compares the 1uc spectrum to $\frac{2}{3}$ of the bulk spectrum along (a) the ($\zeta\pi$, 0) and (b) the ($\zeta\pi$, $\zeta\pi$) symmetry directions. The $\frac{2}{3}$ scaling factor results in similar heights of the single magnon peak, which is known to be transverse. This validates our approach and shows that the domains of locally correlated spins are indeed isotropic and not fixed in the plane. This allows us to distinguish transverse and longitudinal components, leading to two important observations: i) A large component of the continuum is present in both data-sets, meaning that a large component is transverse in nature and therefore cannot be described as 2-magnon scattering in LSWT. ii) At high energy transfers the 1uc shows additional scattering, which must be longitudinal in nature. Unlike the continuum response measured in the bulk film, we observe no clear differences between (0,0)$\rightarrow$(π,0) and (0,0)$\rightarrow$($\frac{\pi}{2}$,$\frac{\pi}{2}$) for this additional scattering, which disperses to higher energies at higher $\boldsymbol{Q}$. In a LSWT picture, such additional longitudinal scattering would come from two-magnon $S^{zz}(\boldsymbol{Q},\omega)$ excitations. These are calculated for La_2CuO_4 (see methods) and plotted for comparison in Fig. 4. We see that the two-magnon response does not accurately account for the additional scattering in the 1uc magnetic excitation spectra.

The ratio of $\boldsymbol{Q}$-integrated longitudinal to transverse scattering can be compared to calculations within LSWT [28]. For a Heisenberg magnet, this ratio is controlled by the spin reduction due to zero-point fluctuations $\Delta S = S - \langle S^z \rangle$, where $\Delta S = 0.2$ for $S = \frac{1}{2}$ and then

$$\frac{\text{Longitudinal}}{\text{Transverse}} = \frac{\Delta S(\Delta S + 1)}{(S - \Delta S)(2\Delta S + 1)} \approx 0.6. \qquad (1)$$

We find that the mean weight of the additional scattering relative to the transverse scattering at the measured $\boldsymbol{Q}$ values is about 0.6, consistent with this estimate, though we note that our measured $\boldsymbol{Q}$ does not constitute a full integration over the zone.

The comparision of bulk and 1uc La_2CuO_4 have revealed a longitudinal and transverse continuum in the magnetic excitation spectrum of La_2CuO_4, which has yet to be fully explained. Possibilities include transverse incoherent scattering [9], higher-order magnons [24, 25] and the presence of proposed [10] spinon-like excitations in La_2CuO_4 [6]. Alongside this, the clearly defined magnon dispersion in 1uc LCO leads to the conclusion that even in the absence of long range order, the ground state hosts classical correlations rather than RVB-like quantum disorder. Further theoretical and experimental studies are called for to explain our observations, and to determine whether these non-LSWT features gain importance upon doping and whether they are relevant to the mechanism of superconductivity in doped cuprates.

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AUTHOR CONTRIBUTIONS

Experiment: M.P.M.D, J.P.H., R.S.S., C.M., K.J.Z., T.S.; Sample growth: I.B.; Sample characterization: I.B., M.P.M.D., R.S.S., A.S., E.M. and T.P.; Two-magnon calculations: B.D.P. and H.M.R.; Data analysis and interpretation: M.P.M.D. J.P.H., J.v.d.B, T.S., C.M., K.J.Z. and H.M.R.; Project planning: J.P.H., M.P.M.D., T.S., I.B.; Paper writing: M.P.M.D and J.P.H., with contributions from all authors.

METHODS

We performed our RIXS experiments at the ADRESS beamline at the Swiss Light Source using the SAXES instrument. The total fluorescence yield at the Cu L_3 edge was measured at regular intervals and the incident energy was tuned to the peak in the absorption. We determined the combined energy resolution of the monochromator and spectrometer by measuring the elastic scattering from carbon tape, which was well described by a Gaussian function with a full width half maximum of 134 meV. The $\boldsymbol{Q}$ resolution was better than 0.004 Å^{-1}. The spectra were normalized to the intensity of the *dd*-excitations in order to account for differing amounts of La_2CuO_4 probed in different films, and to facilitate the comparison of different films for a given scattering geometry. We denote the in-plane scattering vector, $\boldsymbol{Q}$, using the tetragonal La_2CuO_4 unit cell $a = b = 3.8$ Å, with $\boldsymbol{Q}$= (π,0) parallel to the Cu-O-Cu bond direction.

For film synthesis, we employed a unique atomic layer-by-layer molecular-beam epitaxy system equipped with advanced tools for *in-situ* surface analysis including reflection high-energy-electron diffraction (RHEED) and time-of-flight ion-scattering spectroscopy. Using this technique, we reproducibly fabricate single-crystal films with atomically smooth surfaces and interfaces as well as heterostructures and superlattices with superconducting or insulating layers that can be down to one unit cell thick [29, 30]. Digital layer-by-layer growth and the capability to maintain atomic-scale smoothness were both crucial for the present study. The films were grown on single-crystal $LaSrAlO_4$ substrates, each with the 10×10 mm^2 surface polished perpendicular to the (001) direction, under 9×10^{-6} Torr of ozone and at a substrate temperature of about 700 °C. The deposition rates were measured by a quartz crystal oscillator before growth and controlled in real time using a custom-made atomic absorption spectroscopy system. The quality of the film growth was checked by monitoring RHEED intensity oscillations, which provide digital information on the film thickness. The films were subsequently annealed under high vacuum to drive out all the interstitial oxygen and avoid inadvertent oxygen doping. The sample characterization described in supplementary materials [URL to be inserted] shows that these films are a good realization of isolated 1 and 2 unit cell thick La_2CuO_4 layers.

The two magnon excitation spectrum $S^{zz}(\boldsymbol{Q}, \omega)$ was calculated following Ref. [21] for a Hubbard model relevant to La_2CuO_4. The first, second, and third nearest-neighbour hopping parameters were $t = 492$ meV, $t'' = -207$ meV and $t''' = -45$ meV respectively [21] as determined by fitting to neutron scattering measurements [6]. The Coulomb repulsion was fixed at $U = 3.5$ eV .

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